

# Scientific and Engineering Progress in CO<sub>2</sub> Mineralization Using Industrial Waste and Natural Minerals

Heping Xie<sup>1,2\*</sup>, Hairong Yue<sup>3</sup>, Jiahua Zhu<sup>3</sup>, Bin Liang<sup>1,3</sup>, Chun Li<sup>3</sup>, Yufei Wang<sup>1,2</sup>, Lingzhi Xie<sup>1</sup>, Xiangge Zhou<sup>4</sup>

**ABSTRACT** The issues of reducing CO<sub>2</sub> levels in the atmosphere, sustainably utilizing natural mineral resources, and dealing with industrial waste offer challenging opportunities for sustainable development in energy and the environment. The latest advances in CO<sub>2</sub> mineralization technology involving natural minerals and industrial waste are summarized in this paper, with great emphasis on the advancement of fundamental science, economic evaluation, and engineering applications. We discuss several leading large-scale CO<sub>2</sub> mineralization methodologies from a technical and engineering-science perspective. For each technology option, we give an overview of the technical parameters, reaction pathway, reactivity, procedural scheme, and laboratorial and pilot devices. Furthermore, we present a discussion of each technology based on experimental results and the literature. Finally, current gaps in knowledge are identified in the conclusion, and an overview of the challenges and opportunities for future research in this field is provided.

**KEYWORDS** CO<sub>2</sub> mineralization, natural mineral, industrial waste, science and engineering

## 1 Introduction

Carbon dioxide (CO<sub>2</sub>) is the primary greenhouse gas emitted by human activities [1]. According to the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report of Climate Change 2013 (AR5), the atmospheric concentration of CO<sub>2</sub> rose to  $3.91 \times 10^8 \mu\text{mol} \cdot \text{mol}^{-1}$  in 2011—more than 40% higher than the pre-industrial level. A significant share of the total CO<sub>2</sub> sequestration required for climate change mitigation (about 25% of the total in International Energy Agency (IEA) and IPCC scenarios) is expected to result from carbon capture and storage (CCS). CCS has been intensively inves-

tigated in the past couple of decades [2–4]. Potential storage methods include geological storage (storage in oil and gas fields, deep saline formations, or in un-minable coal beds) and ocean storage [5–9]. Current CCS methods are still far from commercialization, due to their high cost and high energy consumption. The storage of CO<sub>2</sub> in the subsurface may involve risks such as leakage, contamination of underground water, or even geological disasters [10, 11].

CO<sub>2</sub> capture and utilization (CCU) methodology is currently more attractive than CCS as the currently available technology for climate change mitigation. Because CCU produces high-value chemical products, it can be used to reduce CO<sub>2</sub> emissions with relatively low overall cost and energy expenditure. However, CO<sub>2</sub> is considered one of the most stable chemical compounds in the carbon family. Although many publications and much research concern the utilization of CO<sub>2</sub> to produce chemicals, mineralization is one of the most efficient methodologies, since it is thermodynamically favorable (Figure 1) [12]. Other methods, such as the conversion of CO<sub>2</sub> to fuel and organic chemicals, require more energy than they produce and theoretically generate more CO<sub>2</sub>.

CO<sub>2</sub> mineralization can store CO<sub>2</sub> in a more stable form for thousands of years via reaction with alkaline earth oxides to form carbonates. Some minerals, such as those in volcanic ash and industrial waste, contain basic alkaline earth compounds, which can be reacted with CO<sub>2</sub>. In recent years, natural minerals such as serpentine and olivine have been extensively investigated for their ability to fix CO<sub>2</sub> by mineralization [13], and MgO- or CaO-containing silicate ores have been thermally activated and reacted with CO<sub>2</sub>. However, activation and mineralization by these methods require a high energy input and have a high cost, and these factors are the major obstacles for the commercialization of these technologies.

<sup>1</sup> Center of CCUS and CO<sub>2</sub> Mineralization and Utilization, Sichuan University, Chengdu 610065, China; <sup>2</sup> College of Water Resources & Hydropower, Sichuan University, Chengdu 610065, China; <sup>3</sup> College of Chemical Engineering, Sichuan University, Chengdu 610065, China; <sup>4</sup> School of Chemistry, Sichuan University, Chengdu 610065, China

\* Correspondence author. E-mail: Xiehp@scu.edu.cn

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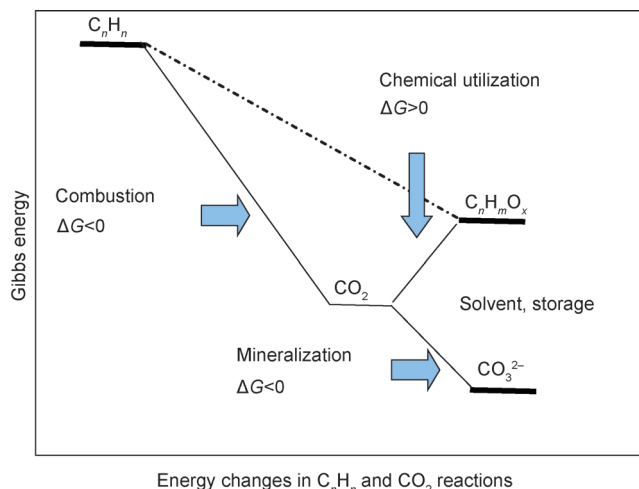


Figure 1. Energy changes in  $\text{CO}_2$  reactions.

Sichuan University has developed a series of technologies to sequester and utilize  $\text{CO}_2$  by mineralization. These technologies have low energy input and are economically viable. Theoretically, the carbonation of  $\text{CO}_2$  with  $\text{CaO}$ - or  $\text{MgO}$ -containing minerals releases energy, which can be recovered. In addition, this process utilizes  $\text{CO}_2$  to produce valuable chemicals. Energy can be recovered during the mineralization of  $\text{CO}_2$ , and used to produce desired chemicals using their chemical potentials.

Figure 2 shows material fluxes and process steps for  $\text{CO}_2$  mineralization. This process is coupled with industrial waste conversion, mineral processing, and energy recovery. Phosphogypsum (PG) is a large-scale industrial waste from the production of wet phosphoric acid [14]. The phosphoric acid industry in China deposits about  $5 \times 10^7$  t of PG every year, resulting in a severe environmental challenge.  $\text{CO}_2$  mineralization was used to convert the PG to  $\text{CaCO}_3$  and  $(\text{NH}_4)_2\text{SO}_4$ , which can be used as a construction material and a fertilizer, respectively. Steel slag containing  $\text{CaO}$  or portlandite ( $\text{Ca}(\text{OH})_2$ ) is an industrial waste from steel plants. This material is a toxic deposit due to the leachable  $\text{CaO}$ . Steel slag is used to produce construction material, and can be used to generate power by  $\text{CO}_2$  mineralization (converting  $\text{CaO}$  to  $\text{CaCO}_3$ ).  $\text{CO}_2$  mineralization was also applied to potassium extraction from K-feldspar ore. In this process, the  $\text{K}^+$  ions were first exchanged with the  $\text{Ca}^{2+}$  ions and then the  $\text{Ca}^{2+}$  ions were fixed by reacting it with  $\text{CO}_2$  [15]. In addition, an electrolytic method was proposed to intensify the mineralization of  $\text{CO}_2$  with earth-abundant natural magnesium chloride from salt lakes in order to recover valuable magnesium carbonate. This method can potentially mineralize as much as several billion tons of  $\text{CO}_2$  [16].

In this work, we describe several representative  $\text{CO}_2$ -mineralization routes in which natural minerals and industrial waste were used as reaction feedstock/components to produce useful chemicals. We summarize various intensification methodologies, such as the electrochemical method [18], thermal activation [15, 19], and the use of additives to enhance the  $\text{CO}_2$ -mineralization reactions [16], with emphasis on scientific progress, engineering

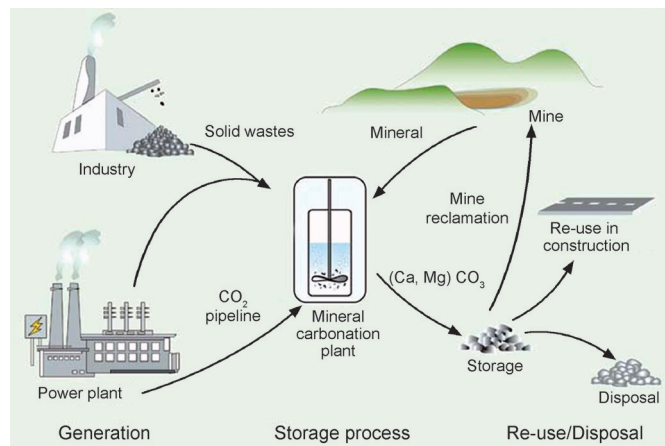


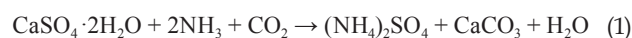
Figure 2. Material fluxes and process steps associated with the  $\text{CO}_2$  mineralization of natural rocks or industrial residues. (Adapted with permission from Ref. [17]. Copyright 2005 Cambridge University Press)

applications, and an economic evaluation of these alternative technologies.

## 2 $\text{CO}_2$ mineralization using industrial waste

### 2.1 $\text{CO}_2$ mineralization with PG for fertilizer production

PG is a waste deposit from the wet-process phosphoric acid industry. China is the largest manufacturer of phosphorous fertilizer and produces  $5 \times 10^7$  t PG per year [14]. However, only 15% of this material is used as a set retarder of cement or to make gypsum plaster and bricks [14, 20]. The rest—a huge quantity of PG—is mostly discarded in large stockpiles without proper disposal, resulting in land occupation and environmental pollution, particularly to water resources. PG can be converted into a nitrogen fertilizer through the following chemical reaction, which can be used in CCU:



The  $\text{CO}_2$  contained in flue gas was captured and converted by ammonium-saturated PG slurry to produce  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{CaCO}_3$  as per Eq. (1). As mentioned earlier,  $(\text{NH}_4)_2\text{SO}_4$  is a fertilizer containing both the nutritious elements for plants, nitrogen (N) and sulfur (S), and  $\text{CaCO}_3$  is a feedstock of cement. This new mineralization approach converted an industrial waste and  $\text{CO}_2$  into two useful products. It is a thermody-

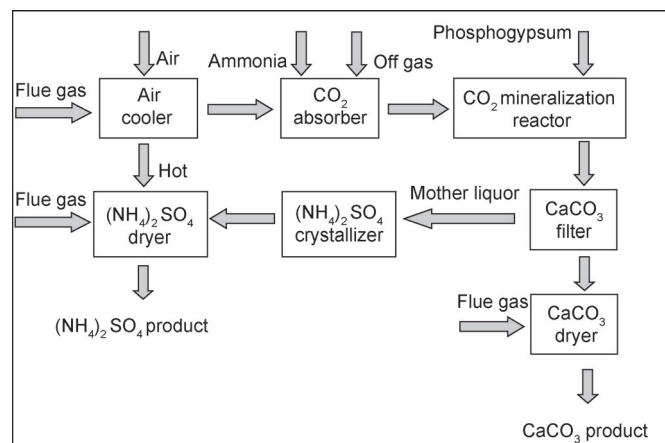
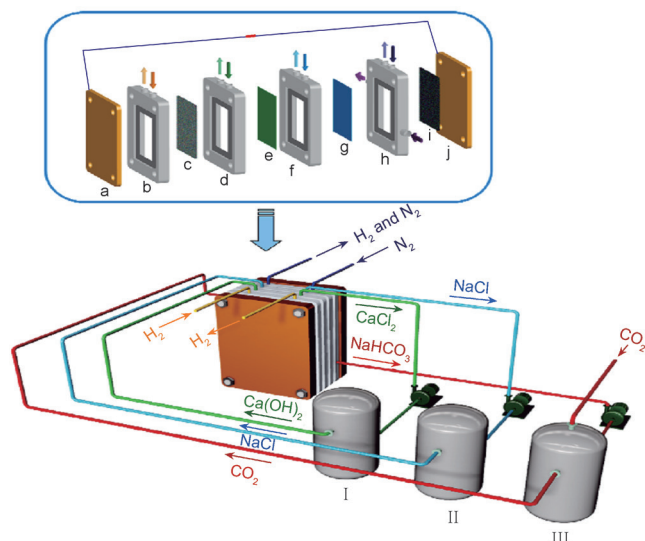


Figure 3. Schematic pilot process for the mineralization of  $\text{CO}_2$  using PG.







**Figure 6. Schematic diagram of the CMFC system and the inner structure of a single CMFC.** (a) Anode current collector; (b) anode gas chamber frame; (c) hydrogen-diffusion anode; (d) anode liquid chamber frame; (e) AEM; (f) salt chamber frame; (g) CEM; (h) cathode chamber frame; (i) cathode; (j) cathode current collector. (I) Anode buffer tank. (II) Salt buffer tank. (III) Cathode buffer tank. (Reproduced with permission from Ref. [22]. Copyright 2014 Science China Press)

process in the  $\text{CO}_2$ -mineralized fuel cell was chosen to enhance the  $\text{CO}_2$  utilization.

In this system, the highly valuable product, industrial  $\text{NaHCO}_3$ , was produced concomitantly during electricity generation. The highest power density of this system was  $5.5 \text{ W} \cdot \text{m}^{-2}$ , higher than that of many microbial fuel cells. The maximum open-circuit voltage was 0.452 V. Moreover, this system was demonstrated to be viable for low concentrations of  $\text{CO}_2$  (10%) and for other carbonation processes. Therefore, this new strategy is an energy-generating and environmentally friendly approach to utilize  $\text{CO}_2$ , and could be used as a supplement to current  $\text{CO}_2$ -emission scenarios.

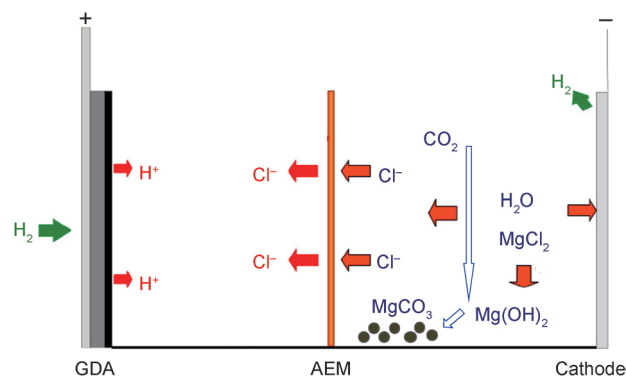
### 3 $\text{CO}_2$ mineralization using natural minerals

#### 3.1 Electrolytic $\text{CO}_2$ mineralization with $\text{MgCl}_2$ to produce $\text{MgCO}_3$

Magnesium chloride ( $\text{MgCl}_2$ ) is an abundant natural resource that is widely distributed in seawater, salt lakes, and ore minerals. The average concentration of magnesium ions in seawater is about 0.13%, which can potentially mineralize as much as  $3.34 \times 10^9 \text{ t}$  of  $\text{CO}_2$  (111 years' worth of global  $\text{CO}_2$  emissions). The prospective reserves of magnesium chloride salt in the four major saline lakes in China are estimated to be as high as several billion tons.

An electrolytic method was proposed to intensify the mineralization reaction of flue-gas  $\text{CO}_2$  with magnesium chloride ( $0.1 \text{ mol} \cdot \text{L}^{-1}$ ) to recover valuable magnesium carbonate ( $\text{MgCO}_3$ ) (Figure 7) [18]. First, magnesium chloride was converted into magnesium hydroxide and hydrochloric acid by electrolysis. Next, the resulting magnesium hydroxide was reacted with  $\text{CO}_2$  to produce a magnesium bicarbonate solution. The magnesium carbonate product was finally obtained by calcination of the  $\text{Mg}(\text{HCO}_3)_2$ .

In the electrolysis cell,  $\text{H}_2$  is oxidized to form  $\text{H}^+$  ions with the help of a special nickel-foil anode. The  $\text{H}^+$  ions react with



**Figure 7. Illustration of an electrolysis cell and the process for the conversion of magnesium chloride to magnesium carbonate.** (Reproduced with permission from Ref. [18]. Copyright 2014 Springer)

the  $\text{Cl}^-$  ions diffusing through the AEM membrane from the cathode side of the cell to form  $\text{HCl}$ . Water is electrolyzed on the cathode and transformed into  $\text{H}_2$ , which is recycled to the anode.  $\text{CO}_2$  mineralization occurs on the cathode side, where the  $\text{Mg}(\text{OH})_2$  reacts with  $\text{CO}_2$  to form  $\text{MgCO}_3$ .

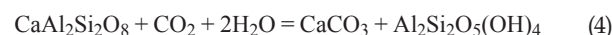
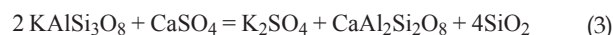
Both pure and diluted  $\text{CO}_2$  gas can be used directly in this mineralization process.  $\text{CO}_2$  concentration can be reduced to below 20%, which is a normal concentration for industrial flue gas. This mineralization method avoids the high-energy-consuming process of  $\text{CO}_2$  purification and greatly reduces the energy consumption of  $\text{CO}_2$  mineralization. About 871 kW·h (about \$70 USD, according to the price of electricity on March 16, 2015) of electricity at a cell voltage of 0.7 V is required to mineralize 1 t of  $\text{CO}_2$  and produce 3.16 t of  $\text{MgCO}_3$ . According to the market price of magnesium carbonate (\$480 USD·t<sup>-1</sup>, according to the price from the CHEMinfo website (<http://www.cheminfo.cn/>) on March 10, 2015),  $\text{CO}_2$  mineralization by means of magnesium chloride is an efficient (net effect of 50%–70%) and economical way to reduce  $\text{CO}_2$  emissions.

#### 3.2 $\text{CO}_2$ mineralization with natural K-feldspar to produce potash fertilizer

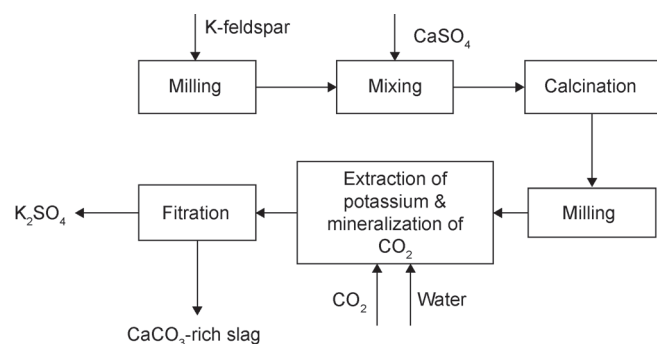
Soluble potassium resources are scarce in many parts of the world (< 1% of global potassium storage), including China. On the other hand, K-feldspar ( $\text{KAlSi}_3\text{O}_8$ ) is an abundant and stable insoluble potash ore with reserves of more than  $1.0 \times 10^{10} \text{ t}$ ; therefore, the production of potash fertilizer from K-feldspar is crucial in order to reduce the consumption of soluble potassium resources [23].

##### 3.2.1 K-feldspar-phosphogypsum co-activation and mineralization process

A coupling process (Figure 8) comprising the activation of K-feldspar with PG at high temperature was investigated as a method of extracting  $\text{K}_2\text{SO}_4$  and mineralizing  $\text{CO}_2$ . The main reactions in this activation and mineralization process are:



This process extracts potassium from natural K-feldspar ore and simultaneously deals with PG waste [15]. The K-extraction and  $\text{CO}_2$ -mineralization rates were significantly affected by activation parameters. Under specific activating



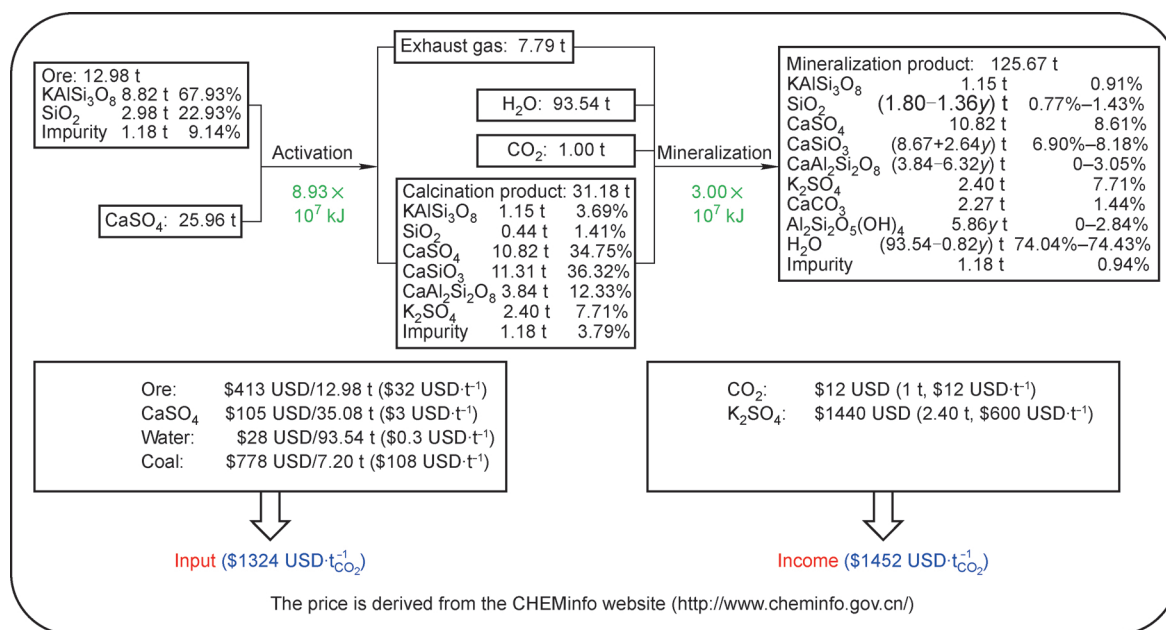
**Figure 8. Schematic diagram of the thermal activation of K-feldspar and PG for CO<sub>2</sub> mineralization.** (Reproduced with permission from Ref. [15]. Copyright 2014 American Chemical Society)

conditions—1200 °C for 2 h with an ore/CaSO<sub>4</sub> mass ratio of 1 : 2, and mineralizing at 100 °C with an initial CO<sub>2</sub> pressure of 4 MPa—the K-extraction and CO<sub>2</sub>-mineralization yields exceeded 87% and 7.7%, respectively.

This potassium-extraction process follows an ion exchange reaction of Ca<sup>2+</sup> from CaSO<sub>4</sub> with K<sup>+</sup> from K-feldspar, in which CO<sub>2</sub> reacts with CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>. The K<sup>+</sup> ions exchange with Ca<sup>2+</sup> ions to form K<sub>2</sub>SO<sub>4</sub>, and the remaining molecular skeleton after this ion exchange forms an electrical neutral phase CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>. The CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> is then dissolved by H<sup>+</sup>, which results from the ionization of H<sub>2</sub>CO<sub>3</sub>, and Ca<sup>2+</sup> is released. Finally, the Ca<sup>2+</sup> is reacted with HCO<sub>3</sub><sup>-</sup> to form CaCO<sub>3</sub> [24].

The reaction activation temperature of 1200 °C is much lower than that of the decomposition temperature of K-feldspar (1500 °C), which significantly reduces the energy consumption of this process. The reaction temperature is expected to be in the range between the particle surface melting (half-melting) and the melting point, in order to maintain the fluidity of the reactants in commercial production. The co-activation of K-feldspar with PG facilitates the formation of a molten phase in the calcination reaction, which enhances the mass transfer and reaction rate and reduces the reaction temperature. Therefore, this process is a viable alternative approach for the application of natural K-feldspar and industrial solid waste, with a relatively low energy consumption.

To evaluate the economic efficiency of this co-activation and mineralization process, the primary balance of the materials and energy needed for the mineralization of one ton of CO<sub>2</sub> was calculated based on the experimental data (Figure 9, *y* denotes the amount of CO<sub>2</sub> mineralized by K-feldspar). The results showed that the primary cost is the input of ore and the energy consumption, while the primary income is the production of K<sub>2</sub>SO<sub>4</sub>. According to the CHEMinfo website, the present market value of K<sub>2</sub>SO<sub>4</sub> is about \$600 USD · t<sup>-1</sup>, so the net profit for this mineralization technology is \$200 USD · t<sup>-1</sup> of CO<sub>2</sub>. Therefore, this coupled activation and mineralization process is an economically feasible method to reduce CO<sub>2</sub> emissions.



**Figure 9. An economic analysis of the K-feldspar-phosphogypsum co-activation and mineralization process, based on the mineralization of 1 t of CO<sub>2</sub>.**

### 3.2.2 K-feldspar-CaCl<sub>2</sub> co-activation and mineralization process

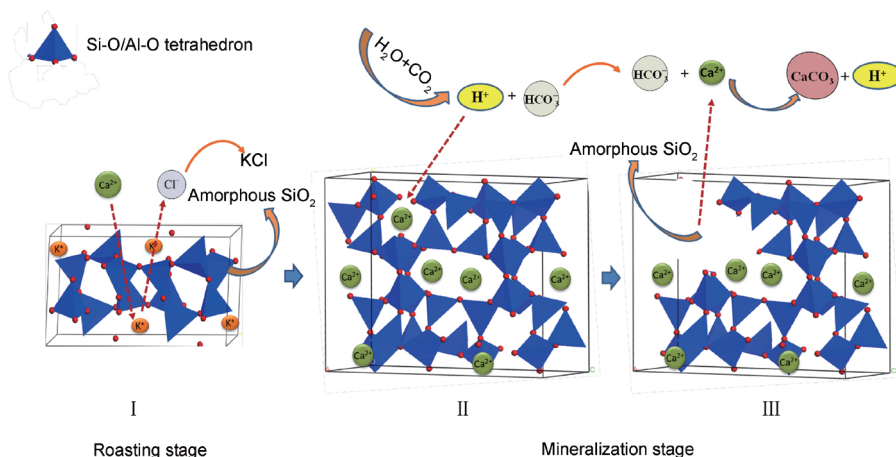
In order to further reduce the activation temperature and the energy consumption, a compound with a lower melting point, CaCl<sub>2</sub> (ca. 782 °C), was used as the promoter for the thermal activation of K-feldspar and mineralization of CO<sub>2</sub> to extract soluble potassium [19]. CaCl<sub>2</sub> is an industrial waste with limited practical use, deposited by the Solvay process of soda-ash production. Therefore, this new process is environ-

mentally friendly, due to its consumption of the CaCl<sub>2</sub> waste.

Activation temperature, reaction time, and the K-feldspar/CaCl<sub>2</sub> ratio are the important factors in this K-extraction and CO<sub>2</sub>-mineralization system. A fairly high K-extraction ratio (over 90%) and CO<sub>2</sub>-mineralization ratio (0.12 gCO<sub>2</sub>/g<sub>K-feldspar</sub>) were obtained at an activation temperature of 908.3 °C—a much lower temperature than that of the PG co-activation system. As shown in the reaction model (Figure 10) the use of an appropriate temperature and CaCl<sub>2</sub> content fa-

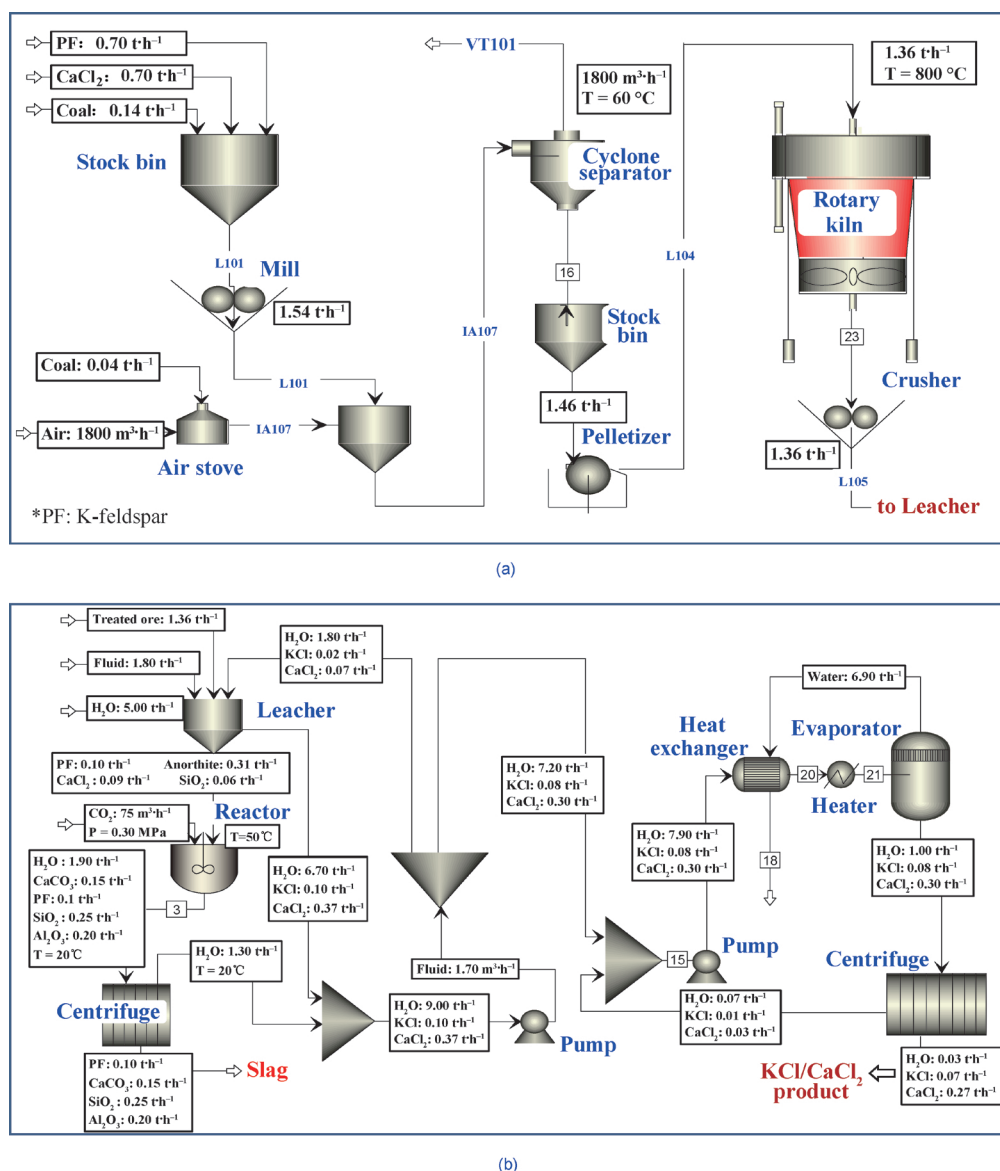
cilitates the exchange of  $\text{Ca}^{2+}$  with  $\text{K}^+$  and the formation of  $\text{CaAl}_2\text{Si}_2\text{O}_8$ ,  $\text{Ca}_3\text{Si}_3\text{O}_9$ , and  $\text{CaSiO}_3$ , due to the hydrolysis of  $\text{CaCl}_2$  and the collapse of the K-feldspar framework.  $\text{CO}_2$  is fixed by reactions with  $\text{Ca}_3\text{Si}_3\text{O}_9$  and  $\text{CaSiO}_3$ , resulting in the mineralization product,  $\text{CaCO}_3$ . The results also showed that the pH of the solution was a significant factor for  $\text{CO}_2$  mineralization. Low pH accelerates the leach of  $\text{Ca}^{2+}$  but restrains the absorption of  $\text{CO}_2$ , while high pH increases the concentration of  $\text{CO}_3^{2-}$ . Therefore, an appropriate pH,  $\text{CO}_3^{2-}$  concentration, and  $\text{Ca}^{2+}$  concentration are key factors for the precipitation of  $\text{CaCO}_3$  in this mineralization reaction.

The results indicate that this alternative technology utilizing insoluble K-



**Figure 10. Schematic diagram of the mechanism of the activation and mineralization stage.**  
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feldspar,  $\text{CaCl}_2$  industrial waste, and  $\text{CO}_2$  mineralization is a promising process with the potential to produce potash fertilizer and reduce  $\text{CO}_2$  emissions.



**Figure 11. The technological process for mineralization of 500 t·a<sup>-1</sup> CO<sub>2</sub> with K-feldspar.** (a) Pretreatment and activation section; (b) leaching and mineralization section.

### 3.2.3 The technological process for mineralization of 500 t·a<sup>-1</sup> CO<sub>2</sub> with K-feldspar

To evaluate the potential of this mineralization technology for industrial application, a scale-up pilot plant was designed on the basis of the experimental results with a mineralization ability of 500 t of CO<sub>2</sub> per year. Figure 11 shows the principal process.

This process involves four main steps: pretreatment, activation, leaching, and mineralization. K-feldspar, CaCl<sub>2</sub>, and coal were first mixed and milled to a particle size below 150 μm, and then shaped and activated at an appropriate temperature (e.g., 800–900 °C) in order to exchange K<sup>+</sup> with Ca<sup>2+</sup> ions from anhydrous calcium chloride. The activated slag was leached with hot deionized water (at a temperature of ca. 80 °C) to extract potassium, and then CO<sub>2</sub> was bubbled in the suspension of the residue at 0.3 MPa and 50 °C for 2 h, for mineralization of CO<sub>2</sub> to occur. The slag was centrifuged and filtered after mineralization, and the filtrate and washing solutions were concentrated to obtain the KCl and CaCl<sub>2</sub> products. This pilot plant has been designed and is currently under construction. At this plant, the technological parameters of the process will

be investigated systematically, to develop further understanding and to pave the way for industrial applications.

## 4 Conclusions and future scope

In recent years, Sichuan University has made remarkable progress in the development of CO<sub>2</sub>-mineralization technologies with high efficiency, low cost, and low energy consumption. We summarize the reactions, relative reactivity, and economic viability for the representative CO<sub>2</sub>-mineralization technologies in Table 1. Based on the cost analysis of these performed processes, the presented CCU techniques exhibit excellent economic viability and potential for application. In this context, we anticipate that research in CO<sub>2</sub> mineralization will become even more widespread in the near future, particularly for the coupled technologies of natural resource exploitation and industrial waste treatment. The technologies described in this paper have the potential to process large quantities of CO<sub>2</sub> and industrial waste, while producing high-value chemicals.

CO<sub>2</sub> mineralization using natural minerals and industrial

**Table 1. Comparison of the reactions, relative reactivity and the economic viability for the processes.**

	Reactions	K-extraction ratio (%)	CO <sub>2</sub> mineralization	Economic viability
Phosphogypsum	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + 2\text{NH}_3 + \text{CO}_2 \rightarrow (\text{NH}_4)_2\text{SO}_4 + \text{CaCO}_3 + \text{H}_2\text{O}$	—	75%	Income \$17 USD · t <sub>CO<sub>2</sub></sub> <sup>-1</sup>
K-feldspar & Phosphogypsum	$\text{KAlSi}_3\text{O}_8 + \text{CaSO}_4 \rightarrow \text{K}_2\text{SO}_4 + \text{CaAl}_2\text{Si}_2\text{O}_8 + \text{SiO}_2$ $\text{CaAl}_2\text{Si}_2\text{O}_8 + \text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{CaCO}_3 + \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$	87%	7.7%	Income \$200 USD · t <sub>CO<sub>2</sub></sub> <sup>-1</sup>
K-feldspar & industrial CaCl <sub>2</sub>	$2\text{KAlSi}_3\text{O}_8 + \text{CaCl}_2 \rightarrow 2\text{KCl} + \text{CaAl}_2\text{Si}_2\text{O}_8 + 4\text{SiO}_2$ $\text{CaCl}_2(\text{s or l}) + \text{SiO}_2(\text{s}) + \text{H}_2\text{O}(\text{g}) \rightarrow \text{CaSiO}_3(\text{s}) + 2\text{HCl}$ $\text{CaSiO}_3(\text{s}) + \text{CO}_2(\text{g}) \rightarrow \text{CaCO}_3(\text{s}) + \text{SiO}_2(\text{s})$	Over 90%	0.12 g <sub>CO<sub>2</sub></sub> /g <sub>K-feldspar</sub>	—
Magnesium chloride	$\text{MgCl}_2 + \text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{MgCO}_3 + 2\text{HCl}$ (In electrolysis cell)	—	—	Income \$70 USD · t <sub>CO<sub>2</sub></sub> <sup>-1</sup>
Portlandite	$\text{Ca}(\text{OH})_2 + 2\text{NaCl} + 2\text{CO}_2 \rightarrow \text{NaHCO}_3 + \text{CaCl}_2$	—	—	5.5 W·m <sup>-2</sup>

**Notes (components of the natural K-feldspar and the industrial wastes)**

**Phosphogypsum:** CaO (28.98%), (9.84%), Al<sub>2</sub>O<sub>3</sub> (0.08%), Fe<sub>2</sub>O<sub>3</sub> (0.08%), MgO (0.05%), SO<sub>3</sub> (41.5%), Na<sub>2</sub>O (0.05%), K<sub>2</sub>O (0.97%), F (0.14%), P<sub>2</sub>O<sub>5</sub> (0.79%).

**K-feldspar:** K<sub>2</sub>O (8.29%), Al<sub>2</sub>O<sub>3</sub> (18.25%), SiO<sub>2</sub> (58.52%), CaO (1.78%), Na<sub>2</sub>O (2.46%), Fe<sub>2</sub>O<sub>3</sub> (2.23%), others (8.47%).

**Portlandite:** CaO (68.99%), (2.84%), Al<sub>2</sub>O<sub>3</sub> (2.16%), Fe<sub>2</sub>O<sub>3</sub> (0.15%), MgO (0.12%), SO<sub>3</sub> (0.76%), Na<sub>2</sub>O (0.03%), H<sub>2</sub>O (24.85%).

**Industrial CaCl<sub>2</sub>:** NaCl (3.2%), CaCl<sub>2</sub> (60.1%), CaO (14.9%), Fe<sub>2</sub>O<sub>3</sub> (1.55%), MgO (10.2%), Al<sub>2</sub>O<sub>3</sub> (4.25%), SiO<sub>2</sub> (8.5%).

wastes has recognized and well-documented advantages, such as very large capacity, no requirement for post-storage monitoring, and overall exothermic processes. However, a number of significant challenges still exist, and must be faced and addressed. Potential exists for breakthroughs in the development of more efficient technologies and optimal systems that allow for the practical mineralization and utilization of CO<sub>2</sub>. Future research should be encouraged in the following areas.

- (1) At present, these processes are still somewhat expensive and are not fully economically favorable. Efforts should be made to assess large-scale technical feasibility and corresponding energy requirements, and to develop simple and cheap methodologies for CO<sub>2</sub> storage.
- (2) Expanding the advertisement to increase public understanding and support is crucial. Public acceptance might be enhanced by the fact that this storage method is highly verifiable and unquestionably permanent.
- (3) Integration of power generation, mining, the carbon-

ation reaction, the carbonates' disposal, and the associated transport of materials must occur to enable energy needs to be optimized in a site-specific manner.

- (4) Future work in mineral carbonation requires the development of a series of demonstration plants in order to enable a deeper understanding of the mineralization processes for industrial applications.

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## Compliance with ethics guidelines

Heping Xie, Hairong Yue, Jiahua Zhu, Bin Liang, Chun Li, Yufei Wang, Lingzhi Xie, and Xiangge Zhou declare that they



have no conflict of interest or financial conflicts to disclose.

## References

1. C. Y. Tai, W. R. Chen, S. M. Shih. Factors affecting wollastonite carbonation under CO<sub>2</sub> supercritical conditions. *AIChE J.*, 2006, 52(1): 292–299
2. W. Wang, X. Liu, P. Wang, Y. Zheng, M. Wang. Enhancement of CO<sub>2</sub> mineralization in Ca<sup>2+</sup>/Mg<sup>2+</sup>-rich aqueous solutions using insoluble amine. *Ind. Eng. Chem. Res.*, 2013, 52(23): 8028–8033
3. V. Materic, S. I. Smedley. High temperature carbonation of Ca(OH)<sub>2</sub>. *Ind. Eng. Chem. Res.*, 2011, 50(10): 5927–5932
4. G. Grasa, J. C. Abanades, E. J. Anthony. Effect of partial carbonation on the cyclic CaO carbonation reaction. *Ind. Eng. Chem. Res.*, 2009, 48(20): 9090–9096
5. D. Tong, J. P. M. Trusler, D. Vega-Maza. Solubility of CO<sub>2</sub> in aqueous solutions of CaCl<sub>2</sub> or MgCl<sub>2</sub> and in a synthetic formation brine at temperatures up to 423 K and pressures up to 40 MPa. *J. Chem. Eng. Data*, 2013, 58(7): 2116–2124
6. K. K. Godishala, J. S. Sangwai, N. A. Sami, K. Das. Phase stability of semi-clathrate hydrates of carbon dioxide in synthetic sea water. *J. Chem. Eng. Data*, 2013, 58(4): 1062–1067
7. X. Li, E. S. Boek, G. C. Maitland, J. P. M. Trusler. Interfacial tension of (brines + CO<sub>2</sub>): CaCl<sub>2</sub>(aq), MgCl<sub>2</sub>(aq), and Na<sub>2</sub>SO<sub>4</sub>(aq) at temperatures between (343 and 423) K, pressures between (2 and 50) MPa, and molalities of (0.5 to 5) mol·kg<sup>-1</sup>. *J. Chem. Eng. Data*, 2012, 57(5): 1369–1375
8. Z. Sun, M. Fan, M. Argyle. Supported monoethanolamine for CO<sub>2</sub> separation. *Ind. Eng. Chem. Res.*, 2011, 50(19): 11343–11349
9. W. Chaikittisilp, R. Khunsupat, T. T. Chen, C. W. Jones. Poly (allylamine)-mesoporous silica composite materials for CO<sub>2</sub> capture from simulated flue gas or ambient air. *Ind. Eng. Chem. Res.*, 2011, 50(24): 14203–14210
10. S. Holloway, J. M. Pearce, V. L. Hards, T. Ohsumi, J. Gale. Natural emissions of CO<sub>2</sub> from the geosphere and their bearing on the geological storage of carbon dioxide. *Energy*, 2007, 32(7): 1194–1201
11. H. Hassanzadeh, M. Pooladi-Darvish, D. W. Keith. Accelerating CO<sub>2</sub> dissolution in saline aquifers for geological storage — Mechanistic and sensitivity studies. *Energy Fuels*, 2009, 23(6): 3328–3336
12. J. Zhu, et al. Thermodynamics cognizance of CCS and CCU routes for CO<sub>2</sub> Emission Reduction. *J. Sichuan Uni. (Eng. Sci. Ed)*, 2013, 45(5): 1–7 (in Chinese)
13. M. Verduyn, H. Geerlings, G. Mossel, S. Vijayakumari. Review of the various CO<sub>2</sub> mineralization product forms. *Energy Procedia*, 2011, 4: 2885–2892
14. H. Tayibi, M. Choura, F. A. López, F. J. Alguacil, A. López-Delgado. Environmental impact and management of phosphogypsum. *J. Environ. Manage.*, 2009, 90(8): 2377–2386
15. C. Wang, H. Yue, C. Li, B. Liang, J. Zhu, H. Xie. Mineralization of CO<sub>2</sub> using natural K-feldspar and industrial solid waste to produce soluble potassium. *Ind. Eng. Chem. Res.*, 2014, 53(19): 7971–7978
16. H. Xie, et al. Simultaneous mineralization of CO<sub>2</sub> and recovery of soluble potassium using earth-abundant potassium feldspar. *Chin. Sci. Bull.*, 2013, 58(1): 128–132
17. B. Metz, O. Davidson, H. C. de Coninck, M. Loos, L. A. Meyer, eds. *IPCC Special Report on Carbon Dioxide Capture and Storage*. Cambridge: Cambridge University Press, 2005
18. H. Xie, Y. Wang, W. Chu, Y. Ju. Mineralization of flue gas CO<sub>2</sub> with coproduction of valuable magnesium carbonate by means of magnesium chloride. *Chin. Sci. Bull.*, 2014, 59(23): 2882–2889
19. L. Ye, et al. CO<sub>2</sub> mineralization of activated K-feldspar + CaCl<sub>2</sub> slag to fix carbon and produce soluble potash salt. *Ind. Eng. Chem. Res.*, 2014, 53(26): 10557–10565
20. İ. Akin Altun, Y. Sert. Utilization of weathered phosphogypsum as set retarder in Portland cement. *Cement Concr. Res.*, 2004, 34(4): 677–680
21. H. V. M. Hamelers, O. Schaetzle, J. M. Paz-García, P. M. Biesheuvel, C. J. N. Buisman. Harvesting energy from CO<sub>2</sub> emissions. *Environ. Sci. Technol. Lett.*, 2013, 1(1): 31–35
22. H. Xie, et al. Generation of electricity from CO<sub>2</sub> mineralization: Principle and realization. *Sci. China Technol. Sc.*, 2014, 57(12): 2335–2346.
23. K. Huang, X. Meng, G. Wang. Research progress of extracting potassium from potassium feldspar. *Phosphate & Compound Fertilizer*, 2011, 26(5): 16–19
24. I. A. Munz, et al. Mechanisms and rates of plagioclase carbonation reactions. *Geochim. Cosmochim. Acta*, 2012, 77: 27–51